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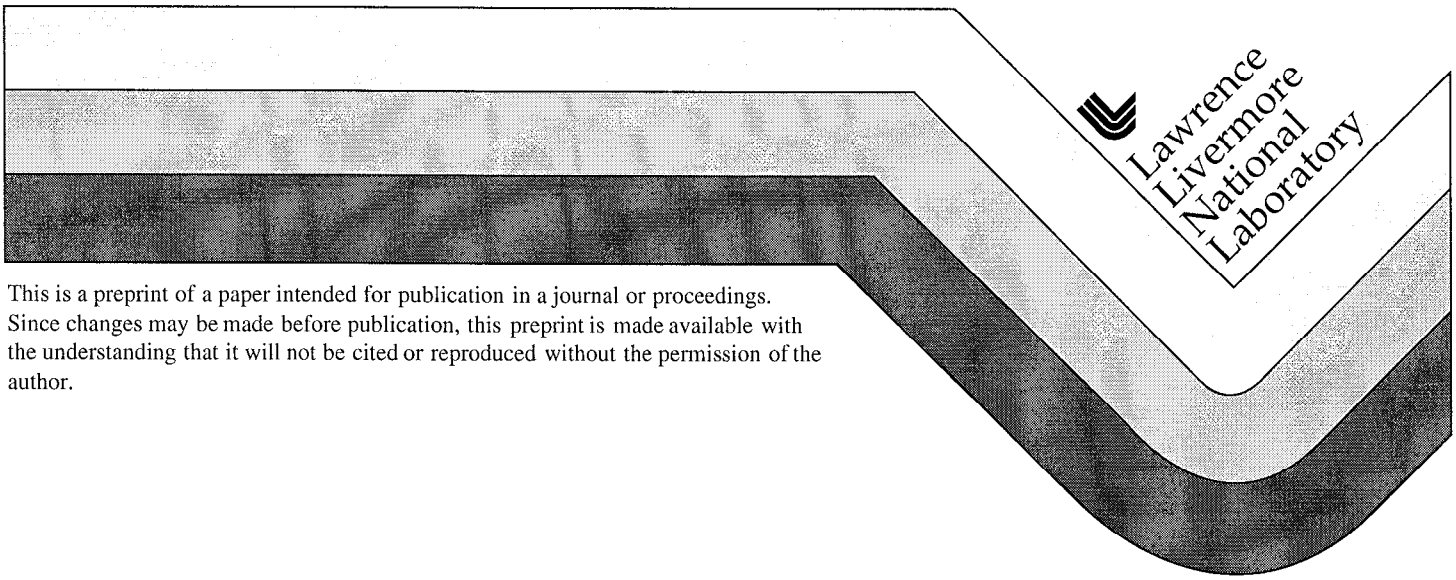

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Elasticity of Tantalum to 105 GPa using a Stress and Angle-resolved X-ray Diffraction

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Determining the mechanical properties such as elastic constants of metals at Mbar pressures has been a difficult task in experiment. Following the development of anisotropic elastic theory by Singh et al. [1], Mao et al.[2] have recently developed a novel experimental technique to determine the elastic constants of Fe by using the stress and energy-dispersive x-ray diffraction (SEX). In this paper, we present an improved complementary technique, stress and angle-resolved x-ray diffraction (SAX), which we have applied to determine the elastic constants of tantalum to 105 GPa. The extrapolation of the tantalum elastic data shows an excellent agreement with the low-pressure ultrasonic data [3]. We also discuss the improvement of this SAX method over the previous SEX.

[elastic constant, anisotropic elastic theory, angle-dispersive synchrotron x-ray diffraction, mechanical properties]

I. Introduction

Elastic constants (C_{ij}) at high pressures and high temperatures are fundamental properties that bridge thermodynamic and mechanical properties and bring an important understanding to material behavior at extreme conditions. For example, C_{ij} measurements of Fe at the condition of the Earth core are extremely valuable to understand the origin of the observed seismic anisotropy of the compression wave velocity of the inner core of the Earth. Recent theoretical calculations of C_{ij} and ideal shear strength of Ta to 10 Mbar [4] and flow stress measurements of Ta over a Mbar [5] certainly widen the interest of measuring elastic constants to higher pressures to understand the behavior of materials at extreme conditions.

Conventional ways of determining elastic constants at high pressures are ultrasonic methods and Brillouin scattering of oriented single crystals. However, these techniques have not established measurements over a Mbar, yet.

Recently, Mao et al. developed a new experimental technique to determine C_{ij} by measuring lattice strains. In this technique, he uses a x-ray transparent Be gasket to reduce the broadening of x-ray peaks and to increase the pressure limit, overcoming the previous limitations [6,7]. Elastic constants of hexagonal close packed (hcp) Fe to over 2 Mbar have been determined using synchrotron x-ray diffraction to aid in our understanding of the interior physics of the Earth [2]. These are the first single crystal elastic constants estimated over a 2 Mbar pressure range. It is important to note that the sample used in this study to determine five single crystal elastic constants of hcp Fe was from a powder sample under uniaxial compression.

However, there are several things to note in this technique. (1) rotation of the diamond cell is required to measure lattice strain as a function of angle (Ψ). (2) this technique used a gold or tunsten as a standard for determination of deviatoric stress of hcp Fe by assuming the axial stress continuity between the sample and the standard layers. This also requires the information of the elastic constants for the standard materials. On the other hand, the angle dispersive x-ray diffraction coupled with an image plate detector that we have developed can improve the quality of the C_{ij} measurements. Our technique, stress and angle-resolved x-ray diffraction (SAX) do not need to rotate the diamond anvil cell (DAC) and the sample is stationary. We also improve our technique without using a standard. We determine deviatoric stress by *in-situ* thickness and pressure gradient measurements.

Deviatoric stress of a sample compressed between the two flat anvils has been estimated to understand the materials strength at high pressures by measuring a pressure gradient and a thickness of a sample. This technique uses a ruby method or x-ray diffraction to measure pressure across a sample. The thickness was measured using an optical microscope for a transparent sample. For metals, the thickness of a recovered sample or gasket was measured to determine the actual thickness during compression. Measuring thickness of a sample after unloading is inevitably the great source of the large uncertainty in the measured properties. We will discuss a new method that we developed to measure in-situ thickness of a sample.

Similar properties can also be estimated by measuring lattice strains with the knowledge of the elastic constants of the standard embedded with sample [6,7]. However, two difficulties have been encountered in those experiments: (1) a broadening of x-ray diffraction line due to a large pressure gradient across an ungasketed sample. (2) an easy failure of diamond anvil during compression limiting the accessible pressure range.

II. Experiments

In the lattice strain measurements using an energy dispersive synchrotron x-ray diffraction, rotation of a DAC can complicates the actual lattice strain measurement. In Fig. 1, we show only two orientations for convenience. Here, with the small 2Θ, the measured strain at $\Psi=90^{\circ}$, for instance can be approximated to represent the lattice strain coming from the lattice planes exactly perpendicular to the axial stress directions, σ_3 [6]. σ_3 is parallel to the compression axis of a DAC. The linear variation of the strain is estimated from the measured d-spacing by rotation of the DAC along the x-ray beam. The angle

dispersive mode we present here does not require rotating a DAC.

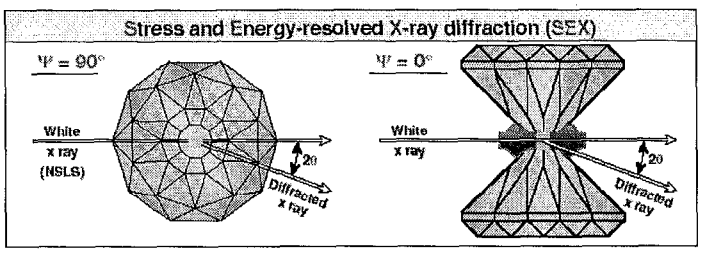

Fig. 1. Radial x-ray diffraction using an energy-dispersive mode. At Ψ=0°, the compression axis of the DAC is tilted by Θ to detect the lattice planes aligned perpendicular to the principal stress axis [2].

Reaching higher pressures in lattice strain measurements using a radial diffraction was achieved by incorporating the x-ray transparent gasket technique [2]. By supporting a powder sample in a cylindrical shape (extremely small diameter, ~ 25 μm) as usual as in a diamond anvil experiment without a pressure medium using an x-ray transparent gasket, the lattice strains from the randomly oriented powder samples are measured as a function of stress angle, Ψ. The angle is defined by the incident x-ray beam and the normal to the diffracting lattice planes (Fig. 2).

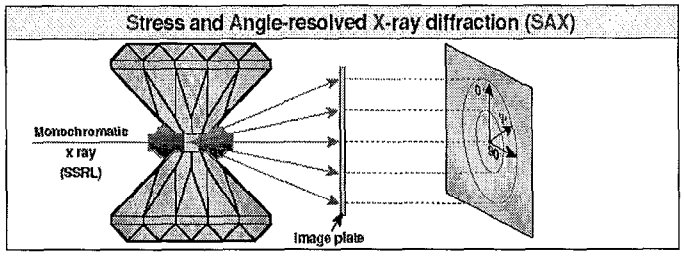

Fig. 2 X-ray diffraction in radial directions using an angle-dispersive mode. The compression axis (σ_3) is perpendicular to the x-ray beam and the image plate is aligned normal to the x-ray beam.

Angle dispersive synchrotron x-ray diffraction using an image plate for the detection of powder diffraction patterns can be developed to measure lattice strains, which differs from the earlier C_{ij} measurements of Fe to 220 GPa using energy dispersive synchrotron x-ray diffraction [2]. In Fig. 1, the stress and energy-resolved x-ray diffraction is shown at Ψ=0° and 90°. The benefit of doing angle dispersive x-ray diffraction using an image plate is apparent since the sample is held stationary unlike the previous energy dispersive x-ray diffraction study, which requires the rotation of the sample along the x-ray beam. Since there is no rotation of the sample, the alignment is simple and the stress angle dependence of the lattice strains is imaged all at once in our method without rotation (Fig. 2).

To accomplish the experimental measurements of the elastic constants of Ta, we have developed a new experimental technique to measure lattice strain. In addition, a new method was developed to measure in-situ thickness of a compressed sample instead of measuring it after unloading as was done previously.[5, 8] Finally, the recently measured equation of state (EOS) of Ta to 174 GPa [9] was used in determining the pressure from the measured volume compression.

Lattice strain by SAX

We measured lattice strain for a Ta powder sample compressed between the diamond anvils and supported by a Be gasket with a 25 μm sample diameter aperture. In order to measure lattice strain using radial x-ray diffraction and an image plate detector, we modified a diamond anvil cell to have an open side-cone with an angle of 90° where the apex is situated at the center of the sample (Fig. 3). The stress and angle-resolved synchrotron x-ray diffraction (SAX) was performed at Beam Line 10-2 in the Stanford Synchrotron Radiation Laboratory (SSRL). A 10 μm diameter collimator was used to align the x-ray optics, DAC, and image plate. The same collimator was used to define the volume of the x-ray beam. To calibrate the sample to image plate distance, two images were taken at two positions along the beam path with the separation exactly known. The sample distance varied slightly each run, but was around 85 cm. We have performed four separate runs in order to extend the pressure ranges. The recent measurements over a Mbar was achieved using beveled anvils of 300 and 100 μm outer and inner rings, respectively. The image plates were exposed between half an hour to an hour and were scanned using a Fuji BAS 2500 scanner with 50 by 50 μm^2 pixel resolution.

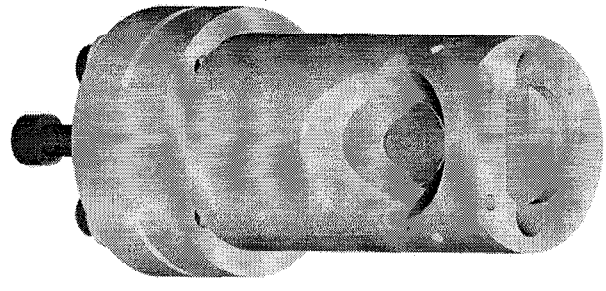

Fig. 3. A modified diamond anvil cell with a side-cone opening and angle cut WC anvil seats to allow measuring lattice strain in the radial direction using angle-resolved synchrotron x-ray diffraction coupled with an image plate up to 2Θ=45°.

Deviatoric stress by x-ray transmission

We also measured deviatoric stress in a separate diamond anvil experiment. We used the same Ta sample for the stress measurements as in the SAX and EOS experiments. The Ta powder was loaded in a Be gasket with an opening of 5 to 10 μm smaller than the culet size of the anvils (300 μm). The sample was initially compressed until the formation of a nice shiny metal luster developed. To avoid spill beyond the aperture area, careful handling and cleaning were exercised. It helps to find the center of the sample during alignment using x-ray transmission intensity. In the experiments taken at the X17c beam line at NSLS, volume was measured across a sample to determine the pressure gradient. The x-ray beam with a dimension of 9 by 6 μm^2 moved across the sample in both the vertical and horizontal directions. The recent EOS data on Ta to 174 GPa were used to obtain the pressure from the measured

volume compression [9]. The deviatoric stress of Ta powder sample can be estimated from the measured pressure gradient and sample thickness [5, 6, 7]. The sample thickness was calculated from the relative x-ray transmission. A foil 10 μm thick was used to calibrate a mass absorption.

Bulk modulus by angle-resolved x-ray

We have used the EOS of Ta to 174 GPa [9]. For comparison, we calculated the room temperature isotherm for tantalum from Hugoniot data [10]. Using the Mie-Grüneisen equation [11] and the zero temperature internal energy calculated from FP-LMTO calculation [4], the experimentally measured values, B_0=194.7 GPa and B_0'=3.4 agree very well with the calculated reduced Hugoniot values, B_0=195 GPa and B_0'=3.6 and the ultrasonic data [3].

III. Data Analysis

We analyzed the experimentally measured lattice strain as a function of the stress angle. As expected, the measured d-spacings of hkl lattice planes shows sinusoidal distribution with two maxima at Ψ=0° and 180° and two minima at Ψ=90° and 270°. The most compressed planes, lying perpendicular to the compression axis, are shown as the minima. The actual position in the image plate is dependent on the way that the SAX-DAC is held, being either in the horizontal or vertical plane defined by the x-ray beam, sample, and the image plate (Fig. 4).

Under uniaxial compression, the powder diffraction patterns become elliptically distorted due to the difference in stress components, σ_1 and σ_3, acting on the randomly oriented lattice planes d(hkl). The elliptical distortion, defined by the ratio of the d-spacing measured under nonhydrostatic (d) and hydrostatic (d_p) conditions, is then a function of the angle Ψ between the diffracting d(hkl) plane and the compression axis σ_3 in Eq (1).

$$d(hkl)/dp(hkl)=1+(1-3\cos^2\Psi)Q(hkl) \quad (1)$$

Q(hkl) represents a lattice strain which depends on the crystal symmetry. For the cubic system [1], it is

$$Q(hkl)=m_0+3m_1\Gamma(hkl) \quad (2)$$

where

$$\Gamma(hkl)=(h^2k^2+k^2l^2+l^2h^2)/(h^2+k^2+l^2)^2 \quad (2a)$$

$$m_0=t(S_{11}-S_{12})/3 \quad (2b)$$

$$m_1=t(S_{11}-S_{12}-S_{44}/2)/3 \quad (2c)$$

Here t is the deviatoric stress, σ_3-σ_1, and S_{11}, S_{12}, S_{44} are the elastic compliances. Eq. (2b, 2c) is simplified based on the isostress approximation across the grain boundaries. Detailed discussions are given in Refs. [1, 2]. For a cubic system [1], the bulk modulus is, $B=1/3(S_{11}+2S_{12}) = (C_{11}+2C_{12})/3$. Using the Eq. 1, the strain parameter Q(hkl) is obtained (Fig. 5). To solve the Eq. (2b and 2c), the estimated deviatoric stress from the measured pressure gradient (Fig. 6) and the thickness is needed.

Our analysis shows that the isostress approximation fits best with the zero pressure data and the pressure derivatives of the elastic constants measured by ultrasonic method [3]. This observation is also confirmed for cubic phases of FeO and Fe [2]. It could be related to the trend in the anisotropy of Ta powder samples, which in fact, remain close to isotropic at high pressure [3]. Looking at the bulk modulus from the collective efforts of SAX-t-EOS measurements appears to have a slightly smaller slope than that obtained from the pressure derivative of the ultrasonic data. However, the measured shear modulus, G seems to be in excellent agreement at zero pressure and at higher pressures compared to the low pressure ultrasonic data [3]. A slight deviation shown in B is also reflected in the C'. In order for our data to match with the zero pressure point from the ultrasonic measurement, a non-linear dependence of the C' seems plausible. On the other hand, C_{44} from the SAX-t-EOS measurements shows excellent agreement with the zero pressure data and also the pressure derivative. Although these two shear moduli apparently show crossing just above 50 GPa in contrary to the near parallel slopes of the moduli in the ultrasonic data, the anisotropy decreases both in our measurements and the ultrasonic data with almost similar slope.

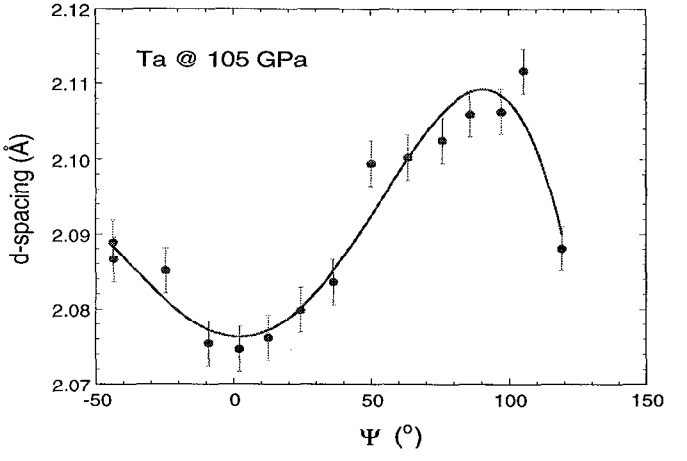

Fig. 4. Plot of the stress angle dependence of (110) d-spacing of Ta powder sample at 105 GPa. The curve is for visual guide only.

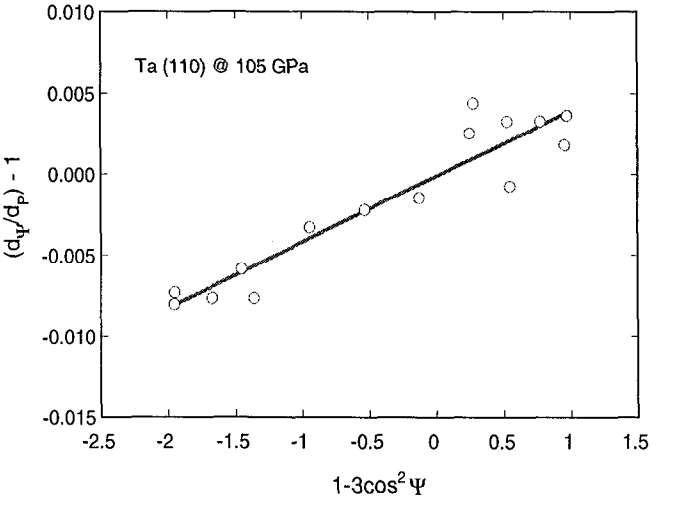

Fig. 5. Plot showing the linear dependence of the strain Q(110) of Ta at 105 GPa as a function of $1-3\cos^2\Psi$. The slope is the strain parameter, Q(110) [Eq. 1].

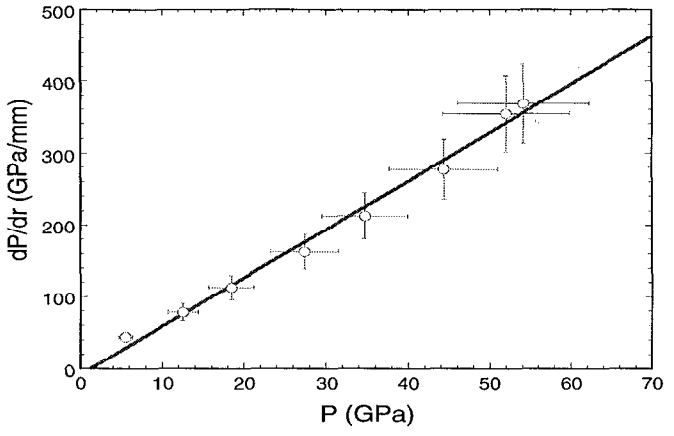

Fig.6. Pressure gradient is shown as a function of the pressure measured at the center of the Ta powder sample uniaxially compressed between two anivls and supported by a Be gasket. The curve is for visual help only.

V. Summary

We have measured C_{ij} values for a Ta powder sample from the integrated efforts to measure EOS, deviatoric stress, and lattice strain (SAX). Our measured values agree very well with the low pressure ultrasonic data both at zero pressure and at higher pressures. This observation confirms the validity of the isostress approximation as seen in the cubic phases of FeO and Fe [2].

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